PATENT ABSTRACTS OF JAPAN

(11)Publication number : 2001-076745
(43)Date of publication of application : 23.03.2001

(51)Int.Cl. H01M 8/02
H01M 8/10

(21)Application number : 11-251373
(22)Date of filing : 06.09.1999

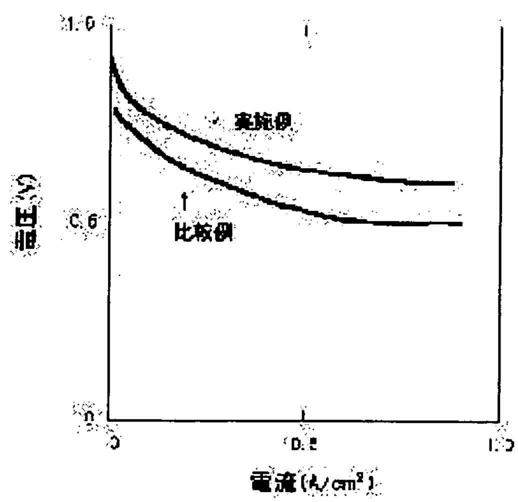
(71)Applicant : TOYOTA CENTRAL RES & DEV LAB INC
(72)Inventor : INOUE KAZUYUKI

(54) CELL OF POLYMER ELECTROLYTE FUEL CELL

(57)Abstract:

PROBLEM TO BE SOLVED: To provide a cell of a polymer electrolyte fuel cell with high energy efficiency and high current-density by improving a polymer electrolyte membrane.

SOLUTION: Insulating granular spacer members are blended into a polymer electrolyte layer interposed between a fuel electrode and an air electrode. The spacer member is a porous ceramic material having moisture-retaining property, such as SiO_2, and blended about 5-25 wt.%. The electrolyte layer can be made as thin as 10-50 μm.



LEGAL STATUS

[Date of request for examination]

[Date of sending the examiner's decision of rejection]

[Kind of final disposal of application other than the examiner's decision of rejection or application converted registration]

[Date of final disposal for application]

[Patent number]

[Date of registration]

[Number of appeal against examiner's decision of rejection]

[Date of requesting appeal against examiner's decision of rejection]

[Date of extinction of right]

DETAILED DESCRIPTION

[Detailed Description of the Invention]
[0001]
[Field of the Invention] This invention relates to the amelioration technique of the polyelectrolyte film interposed between a fuel electrode and an air pole in this kind of fuel cell cel in more detail about a polyelectrolyte mold fuel cell cel.
[0002]
[Description of the Prior Art] Conventionally, the solid-state polyelectrolyte film (only henceforth an "electrolyte membrane") is used as an electrolyte, and since it has the descriptions, like that power density is high, that structure is simple, that operating temperature is comparatively low, and there is silence, this kind of polyelectrolyte mold fuel cell is used as a power source the object for space development, or military. Moreover, when hydrogen gas is used for this fuel cell as fuel gas, since nitrogen oxides and carbon dioxide gas essentially are not discharged, in recent years, it is observed also as a source of low-pollution power for automobiles.
[0003] An example of the basic structure of this polyelectrolyte mold fuel cell is shown in drawing 5 . In drawing 5 , a fuel electrode 34 is joined by one field of an electrolyte membrane 32, and, as for the cell cel 30 used as the generation-of-electrical-energy unit of a polyelectrolyte mold fuel cell, an air pole (not shown) is joined by the field of another side. And it has the structure where inserted with charge collectors (separator) 38 and 38 from the outside of a fuel electrode and an air pole, and the passage 40 of fuel gas, such as hydrogen gas, was formed in the fuel electrode 14 side, and the passage 42 of oxidant gas, such as air, was formed in the air pole side.
[0004] In this case, the fluorine system electrolyte membrane with a thickness of 50-200 micrometers generally represented by the perfluoro sulfonic-acid film known for the trade name of Nafion (a trademark, Du Pont make) is used for said electrolyte membrane 32.
[0005] Moreover, although a fuel electrode 34 and an air pole do not carry out illustration, they are constituted by the two-layer structure where the catalyst bed of the porosity which consists of a carbon particle which made electrode catalysts, such as platinum, support, respectively, and an electrolyte was prepared in the contact surface side with an electrolyte membrane 32, and the diffusion layer which consists of a porous material which can diffuse gas was prepared in the opposite side side.
[0006] As a generation-of-electrical-energy mechanism of the polyelectrolyte mold fuel cell which has such structure If the oxidant gas which contains oxygen, such as air, in a sink and air pole side for the fuel gas which contains hydrogen, such as reformed gas, in a fuel electrode 14 side where a load is connected among 38, the charge collector 38 of the cell cel 30 and is passed Water is generated by the electrochemical reaction from hydrogen and oxygen, and the free energy change in that case is directly taken out from the charge collectors 38 and 38 arranged on the both sides of the cell cel 30 as electrical energy.
[0007] That is, if the reaction conceptual diagram of the fuel cell is shown and explained to drawing 6 , in a fuel electrode 34 side, the fuel gas (hydrogen etc.) supplied through the fuel gas passage 40 serves as a hydrogen ion (H+) and electronic ion (e-), a hydrogen ion (H+) will move in the inside of an

electrolyte membrane 32, it will reach to an air pole, and electronic ion (e-) will be sent to an air pole through a load circuit. And in an air pole 36 side, the oxidant gas (air etc.) supplied to the sent hydrogen ion (H+) and electronic ion (e-), and a list through the oxidant gas passage 42 will react, water (H2O) will be generated, and electrical energy will be taken out by this.
[0008] It is mentioned that the current density of excelling in ** ion conductivity (or proton shift nature), that ** membrane resistance is small, that ** film reinforcement is high, and ** cell is high as a property required of the electrolyte membrane of the polyelectrolyte mold fuel cell of such a configuration, and energy efficiency is good etc. And the attempt which makes an electrolyte membrane thin as much as possible as one of the policies for it is made.
[0009]
[Problem(s) to be Solved by the Invention] However, in the electrolyte membrane of the conventional Nafion system, uniform thickness formation is made difficult. Moreover, film reinforcement is not so enough, either. Therefore, in order to raise the energy efficiency and current density of a cell, when the electrolyte membrane was made thin, in the part with the high pressure which pinches a cell cel with a charge collector (separator), we were anxious about leak of gas occurring from the clearance between an electrolyte membrane and a charge collector.
[0010] Moreover, although it is required for the film itself to be in a moisture state in order for an electrolyte membrane to possess ion conductivity (or proton shift nature), when an electrolyte membrane is made thin, the moisture state will be spoiled. Then, although performing humidification control which adds Myst to fuel gas or oxidant gas, and supplies moisture to an electrolyte membrane is examined in order to maintain the moisture content in an electrolyte membrane, there is also a problem that management of the moisture state of this electrolyte membrane is very difficult.
[0011] The technical problem which is going to solve this invention is to close, if control of the moisture state of an electrolyte membrane is also easy as the moistness of an electrolyte membrane is also maintained further, without raising the film reinforcement of the electrolyte membrane of a polyelectrolyte mold fuel cell cel, and aiming at improvement in the energy efficiency of a cell, and current density by making an electrolyte membrane thin, and an inter-electrode short circuit (short) and an inter-electrode gas leak occurring.
[0012]
[Means for Solving the Problem] In order to solve this technical problem, the polyelectrolyte mold fuel cell cel of this invention makes it a summary to come to blend the spacing member ~~of the shape of a~~ particle which has insulation in the polyelectrolyte layer according to claim ~~1 interposed between a fuel electrode~~ and an air pole like.
[0013] By blending a spacing member with an electrolyte layer, the material strength of an electrolyte layer can be raised, therefore an electrolyte layer can be made thin, and the energy efficiency and current density of a cell can be raised.
[0014] Moreover, since uniform layer thickness is obtained and inter-electrode spacing is also kept constant by the insulating spacing member even if it makes an electrolyte layer thin by raising the material strength of an electrolyte layer therefore, a short circuit (short) arises in inter-electrode, or it is also avoided that the leakage in leak of fuel gas, oxidant gas, etc. occurs from between an electrolyte layer and charge collectors.
[0015] As an ingredient of a spacing member, it is desirable that they are one sort chosen from the oxide system ceramic ingredient and the nitride system ceramic ingredient like or 2 sorts or more of ceramic ingredients ~~according to claim 2,~~ as an oxide system ceramic ingredient, ~~it is mentioned as what~~ has suitable SiO2, aluminum 2O3, etc., and SiN etc. is mentioned as a nitride system ceramic ingredient.
[0016] And even if this spacing member makes an electrolyte layer thin by being the porosity thing according to claim 3 which has moistness like, adsorption maintenance of the moisture for maintaining an electrolyte at a humidification condition will be carried out in the hole of a spacing member etc., and water management can also be performed in a humidification control list for not only the thing that electrolytic proton shift nature is stabilized and is maintained but also an electrolyte to maintain at a humidification condition comparatively comfortably.

[0017] Moreover, as loadings of the spacing member to an electrolyte layer, a thing [that it is in the range of 5 - 25wt%] according to claim 4 is [like] desirable. The water content of an electrolyte layer falls that the loadings of a spacing member are less than [5wt%], and humidification control of an electrolyte becomes difficult. Moreover, if the loadings of a spacing member exceed 25wt(s)%, when an electrolyte layer is compressed with a charge collector, a minute crack etc. will occur in an electrolyte layer and fuel gas and oxidant gas will come to leak to it from there.
[0018] The thinning of an electrolyte layer is attained more sharply than the fuel cell cel for which the thickness of the electrolyte layer of the fuel cell cel of this invention uses a 10-50-micrometer electrolyte membrane by configuration which was mentioned above.
[0019]
[Embodiment of the Invention] The gestalt of suitable operation of this invention is explained with reference to a drawing below at a detail.
[0020] Drawing 1 shows the configuration of the polyelectrolyte mold fuel cell cel concerning 1 operation gestalt of this invention. A fuel electrode 14 is formed in one field into which this polyelectrolyte mold fuel cell cel 10 inserts the polyelectrolyte film 12, and the air pole 16 is formed in the field of another side. And charge collectors (separator) 18a and 18b are arranged in a fuel electrode 14 and air pole 16 side, respectively, and while the fuel gas passage 20 where fuel gas, such as hydrogen gas, flows through is formed in a fuel electrode 14 side, the oxidant gas passage 22 where oxidant gas, such as air, flows through is formed in the air pole 16 side. And this fuel cell cel 10 is assembled in the shape of a laminating, and is used as a laminating mold fuel cell.
[0021] In this case, said polyelectrolyte film 32 uses the particle of silicon oxide (SiO_2) as a spacing member at the polymer of the perfluoro sulfonic acid known for the trade name of Nafion (a trademark, Du Pont make) mentioned already, and what carried out distributed combination is used. Thickness is taken as the range where 10-50 micrometers is suitable.
9/14
[0022] As a configuration of the particle of silicon oxide, the thing of various kinds of configurations, such as the shape of a globular shape, a column, and a rod, is applied. Although especially the particle diameter is not scrupulous, its thing with a diameter of about 10 micrometers is desirable. It can replace with silicon oxide and an alumina (aluminum 2O3), nitriding aluminum (AlN), etc. can also be used. What is necessary is just the thing of the shape of a particle which is insulation, is porosity as it has moistness, and had moderate hardness in short. As loadings of a spacing member, it considers as the range of 5 - 25wt%.
Catalyst
[0023] Moreover, as a fuel electrode 14 and an air pole 16 are expanded to drawing 2 and it was shown, catalyst beds 14a and 16a are formed in a contact surface side with an electrolyte membrane 12, and the gaseous diffusion layers 14b and 16b are formed in the contact surface side with Separators 18a and 18b. Both the catalyst beds 14a and 14b are layers which made the carbon particle support electrode catalysts, such as platinum (Pt), and the gaseous diffusion layers 16a and 16b are constituted by the porous material. For example, the carbon system ingredient of the porosity which the ingredient which can reconcile the diffusibility of matter, such as reactant gas, generation gas, and water, and electronic conductivity, the thing which fabricated carbon paper, a carbon cross, or carbon powder in the shape of a sheet with giant-molecule binders, such as polytetrafluoroethylene, specifically have permeability, and moreover has uniform pore diameter distribution is used. Furthermore, the current collection engine performance of a charge collector (separator) is high, and, generally the graphite of the stable substantia compacta is used also under the oxidation steam ambient atmosphere.
[0024] Next, an example is explained.
The spherical porosity particle with a diameter of 10**0.2 micrometers which measures an equivalent for the polymer content of 40mg, and becomes this solution from silicon oxide about the alcoholic solution (10% of polymer contents, Du Pont) of Nafion (a trade name, trademark of Du Pont) which is ion-exchange resin of a <example> perfluoro sulfonic-acid polymer was added, and it distributed for 20 minutes using the ultrasonic distribution machine of output 200W. It dried using the vacuum dryer, having slushed the solution after distributed processing into the mold of 10cm angle, and imposing the load below the compression breaking load of a silicon oxide particle (for example, 10 g/cm2) on a

solution. The catalyst was applied to the electrolyte membrane with a thickness of 15 micrometers obtained, the gaseous diffusion layer was further pinched with the collecting electrode plate (separator) in piles, and the fuel cell cel was produced. The silicon oxide particle loadings to an electrolyte membrane are made into 15%.
[0025] Moreover, the following sample offering sample was created as an example of a comparison. Using 112 film (50 micrometers in desiccation thickness) of Nafion (same as the above) which does not contain a particle in a <example of comparison> electrolyte membrane, the catalyst was applied to this film, the gaseous diffusion layer was further pinched with the collecting electrode plate (separator) in piles, and the fuel cell cel was produced.
[0026] The current-voltage characteristic of the fuel cell cel of the above-mentioned example when carrying out air at a positive electrode and carrying out 2ata supply of the pure hydrogen at a negative electrode, respectively and the example of a comparison is shown in drawing 3 . In drawing 3 , the current (A/cm2) was taken on the axis of abscissa, and the electrical potential difference (V) is taken on the axis of ordinate.
[0027] And the direction of the fuel cell cel of this example is changing with the current-voltage characteristic higher than the fuel cell cel of the example of a comparison so that the data of this drawing 3 may show. If electrical energy with the fuel cell cel of this example higher than the fuel cell cel of the example of a comparison is obtained and this makes an electrical potential difference the same, it is shown that high current density is obtained. And as a reason this result was obtained, it was large to have made the electrolyte membrane thin too, and the minus by blending a silicon oxide particle with an electrolyte membrane was not accepted.
[0028] Drawing 4 shows the relation between a gas leak incidence rate (%) and the water content (%) of an electrolyte membrane, and the loadings (wt%) of the silicon oxide particle blended with the polyelectrolyte film about the fuel cell cel of this example. In drawing, the loadings (wt%) of a silicon oxide particle were taken on the axis of abscissa, and a gas leak incidence rate (%) and water content (%) are taken on the axis of ordinate.
[0029] and -- as are shown in this drawing 4 , and about 40% of value is shown in the condition (0wt%) of not blending a silicon oxide particle at all, about the introduction gas leak incidence rate (%) and the loadings of a silicon oxide particle are increased -- a gas leak incidence rate -- falling -- about 15 wt(s)% -- the gas leak incidence rate showed 5% and the lowest value in the blended place. And when the loadings of a silicon oxide particle were further increased from this, a gas leak incidence rate tends to worsen and brought a result which falls to 25% of gas leak incidence rates at about 40 wt(s)%.
[0030] From this result, by blending a silicon oxide particle with the polyelectrolyte film shows that a gas leak incidence rate falls and blending a silicon oxide particle with an electrolyte membrane contributes to decline in a gas leak incidence rate. and -- as the loadings of a silicon oxide particle -- about 15wt% -- most -- good -- about 5-25wt% -- when blending, it came out enough and a certain thing became clear.
[0031] Moreover, in the condition (0wt%) of not blending a silicon oxide particle at all, as shown in this drawing 4 , about the water content (%) of an electrolyte membrane, the inclination for water content to rise is shown, hardly including water as the loadings of a silicon oxide particle are increased. And even if water content showed about 40% of value according to about 25wt% and the loadings of a silicon oxide particle increased the loadings of a silicon oxide particle more than this, the rise of water content became loose and brought a result that it seldom changes.
[0032] And it turns out that the water content of an electrolyte membrane rises and the moistness of an electrolyte membrane is raised by blending a silicon oxide particle with the polyelectrolyte film from this result. Since a silicon oxide particle is porosity, it is considered that adsorption maintenance of the moisture will be carried out into the hole of the particle etc. The amount of supply of the water from the outside for this to maintain an electrolyte membrane at a humidification condition can be reduced, and control of the humidification condition becomes easy.
[0033] Alterations various in the range which is not limited to the above-mentioned gestalt of operation at all, and does not deviate from the meaning of this invention are possible for this invention. For

example, although the perfluoro sulfonic-acid polymer was used for the polyelectrolyte film in the above-mentioned example, also when it is presupposed that the ingredient of ****** is excelled most and ingredients other than this are used, it is clearer than the meaning of this invention that this invention is applied. Moreover, although the example of the particle of silicon oxide was explained as a spacing member blended with this, as this was also already described, it cannot be overemphasized that other ceramic ingredients are used.

[0034]

[Effect of the Invention] By blending the spacing member of the shape of a particle which has insulation in the polyelectrolyte layer interposed between a fuel electrode and an air pole according to the polyelectrolyte mold fuel cell cel of this invention Since the material strength of an electrolyte layer is raised and it can be made to make an electrolyte layer thin, even if it makes an electrolyte layer thin by being able to aim at improvement in the energy efficiency of a cell, and current density, and raising the material strength of an electrolyte layer Since thickness of an electrolyte layer can be made into homogeneity, neither an inter-electrode short circuit (short) nor the leakage in leak of gas is produced, and the safety on fuel cell operation is also secured. And when the spacing member blended with an electrolyte layer considers as the porosity thing which has moistness, water management also becomes easy at the humidification control list of an electrolyte layer, and loading as an automotive fuel cell of the practicality etc. is very high.

[Translation done.]